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Diffusion scattering from interstitials in fcc lattices— an optical transform study

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Masks giving the displacements of the atoms surrounding the interstitials have been prepared for several cases in the f.c.c. lattice configurations. Optical diffraction patterns of these masks as well as that of the undistorted lattice have been obtained by using a laser diffractometer. A comparative study of the effect of the displacement field on the intensities and position of the fundamental reflections, and as well as of the modulation of the diffuse scattering intensity in reciprocal space has been made for the above cases.

1. Introduction

Due to the elastic strain around the point defects there is a displacement of the atoms from their normal positions in the lattice. That crystals containing such defects would give rise to diffuse scattering of X-rays as shown by Huang (1947) where a random distribution of the defects each producing a spherically symmetric displacement field $\boldsymbol{U} = c\boldsymbol{r}/|\boldsymbol{r}|^3$ was considered. This theory has been further developed by several workers and with improved experimental techniques it has now been possible to study the relaxation around the lattice defects from diffuse scattering measurements (Krivoglaz 1969, Dederichs 1973). These works show that the Bragg reflections are shifted in positions from those expected from a undistorted lattice, and their intensities are functionally similar to the Debye Waller factor for the thermal vibration of the atoms. But there is no broadening of the reflections due to the defects of this type. Further, there is a diffuse scattering which shows strong directional dependence. However, as discussed by Howard (1971) there is a controversy regarding the exact location of the maxima of the diffuse intensity near the reciprocal lattice points. In the calculation of the diffuse scattering intensities, enormous numerical computations are involved for obtaining satisfactory results and hence often one has to make many simplifying assumptions leading to departures from more realistic models. Moreover, as shown by Keating & Goland (1971), series termination effect in the calculation of the diffuse intensity gives rise to serious errors in the form of ripples in the intensity distribution.

Hence, in the present investigation we have made use of an alternative approach which is free from most of the above difficulties. It is the well known method of the optical transforms in which first a mask is prepared based on realistic models from computer simulation studies, and then the optical diffraction pattern of the mask is taken. A comparison of the X-ray diffuse scattering photographs of a metal with the optical transforms of several such models would help us in getting preliminary information about the type of defects present. Quantitative measurements of the intensities in the different regions of the reciprocal space in the optical transforms would provide a check for the validity of the various theories, thus leading to the exact determination of the anisotropic displacement field around the point defects.

In fact the optical transform studies have been widely used for solving crystal structures (Lipson & Taylor, 1958) and for interpreting X-ray small angle scattering patterns (Hosemann 1962). Willis (1958) has applied optical transform techniques for studying growth faults in cobalt. Diffuse scattering associated with the dislocations has also been studied by Willis (1957). However, no work on its application in the field of Huang scattering from point defects has been reported till now.

2. Choice and Description of the Models

Two of the simplest types of lattice defects are vacant sites and extra atoms wedged in the intersticies of the lattices These two are usually referred to as vacancies and interstitials. Though the location of the vacancies are fixed, the extent of displacement of the neighbouring atoms is uncertain. The situation is more complex in interstitial since the interstitial atom can take up any one of the large number of holes in the lattice thus giving rise to several stable configurations Even one can obtain several such configurations with almost the same interaction energy making it impossible to choose which is the one more stable and more predominent e.g., as shown by Johnson & Brown (1962), the dumbell, the body centered and the crowdion configurations have formation energies 4.0 ± 0.5, 3.9 ± 0.5, and 4.7 ± 0.1 electron volts respectively for copper. Because of the assumptions involved at least ony one of the first two can be the most stable one. Hence a comparative study of the diffuse scattering associated with these configurations would help us in obtaining informations regarding which of those three configurations occur predominantly in different materials. Further because of the close packing of the atoms, the interstitials in the f c c. lattice give rise to large strains and the relaxations are not even approximately spherically symmetric. Hence, a study of the diffuse scattering from above three types of interstitial configurations has been undertaken with the help of an optical diffractometer and the patterns have been compared with that obtained from the undistorted lattice.

In recent years high degree of sophistication has been achieved in the computer simultation study of lattice defects, thus enabling one to obtain a realistic picture. In most of these calculations the lattice is divided into two regions—in the vicinity of the defect where an atomistic picture of the interactions is considered and in the more distant regions an elastic continuum model is assumed. A proper choice of the interaction potential is made and the local distribution of the valance electrons is taken into consideration Finally the equilibrium configurations are obtained by using the variational lattice statistics or Monte Carlo methods. Johnson & Brown (1962) and others have reported such computer simulation studies for interstitial atoms in f.c.c lattices The present models shown in figures 1(a)–1(d) are based on their results.

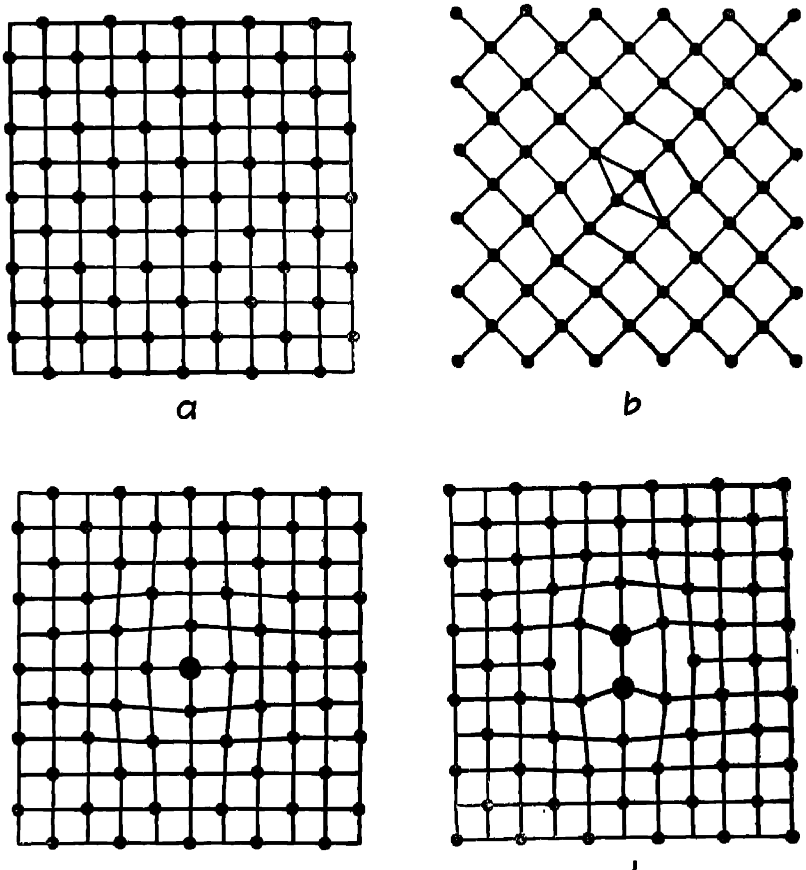


Fig. 1. Models of the normal and distorted f.c.c. lattices containing interstitials—*a*, normal lattice, *b*, crowdion; *c*, body centred; and *d*, dumb-bell configurations.

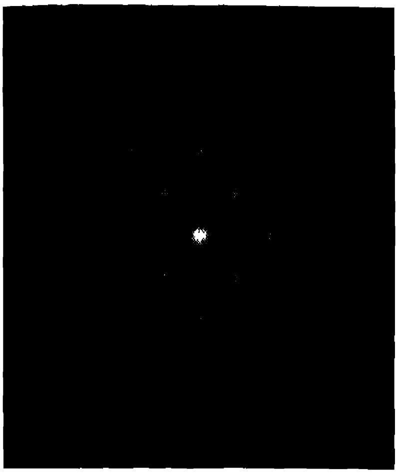
Normal f.c.c. lattice has been shown in figure 1(a). The crowdion configuration has been shown in figure 1(b), where the displacements are large in the < 110 > direction compared to those in other directions. In the body centred configuration (figure 1(c)) the extra atom occupies the largest open space at the centre of the f.c.c. unit cell. In the dumbell configuration (figure 1(b)) the extra atom shares a lattice site with another atom with the axis of the pair lying along the $< 100 >$ direction.

3. Experimental

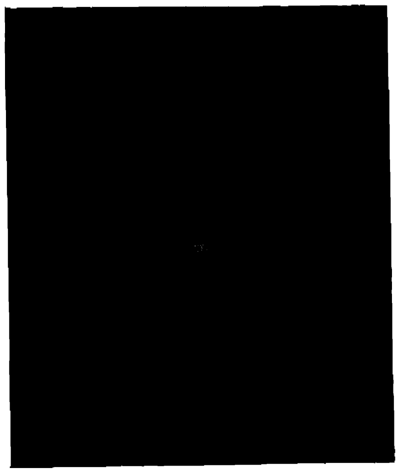
Drawings based on figures 1(a)–1(d) were prepared by plotting 20×20 atoms on a white sheet with an interatomic separation of 2 cm. (in the undistorted case) and photographs were taken with a reduction rate of 100 : 1. In some of the drawings the interstitial atom was drawn of a bigger size for studying the effect of increase in the scattering power on the diffuse intensity. The negatives placed between two optical flats were used as the masks for obtaining the diffraction patterns. An optical diffractomer was constructed with the geometry similar to the well known Lipson diffraction apparatus, but by replacing the conventional Hg-source by a 1 mw spectra-physics He–Ne laser. Experimental details about the construction of the same shall be reported elsewhere. The optical diffraction patterns were recorded on 22 DIN- 125 ASA films and were processed under identical conditions. Figures 2(a)–2(d) are the contact prints of the optical diffraction patterns obtained from the masks corresponding to the figures 1(a)–1(d) respectively.

4. Results and Discussions

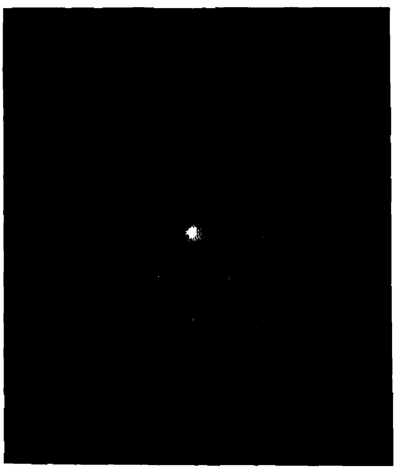
In a crystal of infinite size with atoms rigidly fixed at the lattice points of the periodic lattice, the scattering is given by delta type intensity distribution functions localised at the reciprocal lattice points. Such an intensity distribution was observed in the optical transform of the normal undistorted lattice (figure 2(a)). It consists of a large number of sharp spots and does not exhibit any diffuse scattering. All the transforms, both from normal and distorted lattices, obey the extinction condition characteristic of the f.c.c. lattice, i.e., all the reflections with mixed indices are absent. Compared with the ideal lattice the Bragg reflections are observed to be modified in two ways in the case of the distorted lattices. Firstly there is a minute shift in the positions of the maxima because of the uniform expansion of the lattice. Secondly there is a reduction in the intensities of the reflections. Such a reduction is clearly evident in all the optical transforms of the distorted lattices and is the reason for the absence of high angle reflections in figures 2(b)–2(d). This reduction in intensity is functionally similar to the Debye-Waller factor, $\exp(-2M)$, for the case of dynamical displacement of the atoms due to thermal vibrations. But M in this case is not proportional to q^2, where q is the reciprocal lattice vector. Higher order terms in q comes into play for the case of strong displacement fields. Further as has been discussed by Krivoglaz (1969), M becomes directional dependent. It is observed



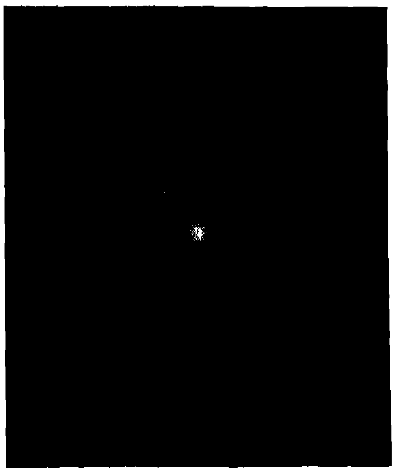
a



b



c



d

Fig. 2. *a, b, c, d*, optical transforms of the models shown in figures 1a, 1b, 1c, 1d respectively.

that the fall in the intensities in the case of optical transforms of the crowdions (figure 2(b)) is much larger compared to that of the body centered and dumbell configurations as shown in figures 2(c)–2(d) respectively. No broadening of the reflections is observed in the present case, though the reflections are known to broaden considerably in case of extended defects.

In addition to the above two cases of changes in the fundamental reflections, occurrence of diffuse scattering is observed in the case of the distorted lattices. The diffuse scattering shows strong directional dependence, this dependence being different for the three types of interstitial configurations. Strong diffuse scattering in the form of cylindrical steaks parallel to the diagonal $< 110 >$ direction is observed in the case of crowdian configuration. Similarly, as shown in figure 2(d), the dumbell interstitical configuration gives rise to broad but extended streaks parallel to the $< 100 >$ direction. However, no such diffuse streaks are observed for the body centred configuration (figure 2(c)). The occurrence of elongated streaks in the crowdion and the dumbell configurations are possibly due to the highly anisotropic displacements in the above directions. The observations are in agreement with the predictions of Kanzaki (1957) where he had predicted that one would get ellipsoidal type diffuse scattering for anisotropic displacements. Thus these quantitative differences in the occurrence of the diffuse intensity streaks can be used to characterise the three types of the configurations. Visual examination of the X-ray photographs for the occurrence of such streaks would indicate which of the three configurations is more predominant for the material.

Due to the displacement of the atoms around the defects one gets a strong diffuse scattering near the reciprocal lattice point. Because of $\boldsymbol{r}/|\boldsymbol{r}|^3$ behaviour of the displacement fields the intensity of the diffuse scattering varies as $1/\boldsymbol{q}'^2$, where $\boldsymbol{q}'$ is the deviation from the reciprocal lattice point. This scattering is known as Huang scattering and has an inversion symmetry around the Bragg reflection. In addition to the Huang scattering one also obtains a scattering which varies as $1/\boldsymbol{q}'$ because of the interaction of the scattering from atoms far from the defect with that from the defect and the highly displaced atoms near the defect. This term is antisymmetric, i.e., it enhances the diffuse scattering on one side of the reflections and diminishes on the other side. Thus for strong displacement fields the intensity is stronger on low angle side of the reflections in case of vacancies, whereas it is stronger on the high angle side in the case of interstitials. Such enhancement of the diffuse intensity on the high angle side is clearly visible in the optical transforms of the distorted lattices specially in figures 2(c) and 2(d). Keating (1968) has shown that such enhancements occur on the high angle side of the Bragg reflection in case of interstitials.

As has been discussed by Dederichs (1973) the diffuse scattering in the region between two reflections is of great importance because it is highly sensitive to the displacements near the defects whereas the Huang scattering is primarily determined by the asymptotic displacement field. Hence a study of the diffuse scattering in the regions away from the Bragg reflections would give informations regarding the exact displacement of the atoms neighbouridg the defects. As has been mentioned earlier the optical transforms from crowdion and the dumb-bell configurations, as shown in figures 2(b) and 2(d), exhibit strong streaks which nearly extend throughout the region between reciprocal lattice points in certain directions. In the crowdion configuration there is a long range relaxation along the $< 100 >$ row which is same as the direction of the strong diffuse streaks observed. Similarly in the dumb-bell configuration the strongly displaced pair of atoms at the defect site have their axis parallel to the $< 100 >$ direction, consequently giving rise to broad streaks in the same direction in the reciprocal space. The contrast in the sharpness of the streaks for the two cases is remarkable. Since the displacement of the atoms in the directions other than the relaxation direction in crowdion is negligible compared to that in the said direction, it gives rise to sharp streaks. In case of the dumb-bell configuration shift in the other directions is considerable even though it is comparatively much smaller than the shift in the $< 100 >$ direction. Hence there is a broadening of the streaks. The displacements of the atoms neighbouring the interestiial in the body centered case is almost uniform in all the directions. Hence its transform does not exhibit any diffuse streak other than the diffuse scattering surrounding the Bragg reflections.

Circles of different radii were placed at the impurity atom position to represent atoms of different scattering factors. This should give rise to monotonous diffuse scattering slowly decreasing with q. But such scattering was too small to be detected.

Thus optical transform study of diffuse scattering from lattice defects can be usefully employed along with X-ray scattering for studying the relaxations produced due to the defects. The authors are carrying out further investigations on the quantitative determination of the displacement fields around the defects by measuring the intensities in their transforms. This would provide a check for the validity of the existing theories of diffuse scattering.

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